

High-T_c Superconductor-Normal-Superconductor Junctions with Polyimide-Passivated Ambient-Temperature Edge Formation

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Abstract Superconducting-Normal-Superconductor (SNS) edge Josephson Junctions (JJ's) continue to be a focus of high-T_c superconductor (HTS) electronics development. The ability to form clean edges in HTS materials is essential for successful SNS edge JJ's as well as crossovers and via contacts. We report a novel SNS JJ fabrication process in which a YBa₂Cu₃O_{7-δ} (YBCO) base electrode/SrTiO₃ (STO) insulator bilayer coated with a polyimide passivation layer is rotated during Ar ion milling through a reflowed photoresist mask to form omnidirectional edges. No sample cooling is used during milling. YBa₂Cu_{2.8}Co_{0.2}O_{7-δ} (YBCCO) normal layers and YLa_{0.05}Ba₂Cu₃O_{7-δ} (YBICO) counter-electrodes completed the SNS JJ's. Devices fabricated using this polyimide passivation layer have significantly smaller standard deviations in critical current and resistance than those processed without it.

1. INTRODUCTION

The ability to controllably fabricate high-temperature Superconductor(Normal-Superconductor (SNS) Josephson Junctions (JJ's) enhances the possibilities for many applications, including digital circuits, SQUID's, and mixers. A wide variety of approaches to fabricating SNS-like junctions has been tried and analyzed in terms of proximity effect behavior[1]. Since their initial successful demonstration[2],[3], most work has focused on SNS edge junctions for circuit applications. In particular, edge SNS JJ's with normal YLa_{0.05}Ba₂Cu₃O_{7-δ} (YBICO) interlayers [4]-[6] are currently favored due to demonstrations of reproducible control of device parameters.

Very recently, high-T_c SNS JJ's on superconducting groundplanes were demonstrated [5], [6]. In order to assure successful fabrication of junctions on groundplanes, separate patterning of the groundplane and insulating oxide layers was eliminated. This minimized the number of layers exposed to photoresist/ion-milling processing and potential degradation of exposed surfaces and subsequently grown films due to

processing residues. While this simplification allowed the very important demonstration of high-T_c SNS JJ's on groundplanes, further improvements in the processing of edges are needed in order to make this technology more flexible.

The use of polyimide films has resulted in higher quality structures and cleaner surfaces in low-temperature superconducting materials processing[7],[8]. To this end, we investigated the use of a polyimide layer to reduce unwanted photoresist residues remaining after processing of oxide surfaces. In this work, we employed the edge SNS junction structure as a test vehicle to determine the relative merits of such a passivation scheme. We show that high-quality SNS devices can indeed be fabricated by including a polyimide passivation layer under the photoresist milling mask and that better control of the device parameters is achieved than in similarly fabricated devices fabricated without such a polyimide layer.

II. JUNCTION FABRICATION

A. Polyimide-Passivated Edge Junctions

SNS edge junction fabrication began with off-axis spotter deposition of YBa₂Cu₃O_{7-δ} (YBCO, 220 nm)/SrTiO₃ (STO, 300 nm) bilayers deposited onto 5 cm-diameter LaAlO₃ (100) wafers. Details of the deposition of these films were discussed previously [5]. The transition temperature, determined by ac susceptibility, of the YBCO film was typically 86-87 K, with a width smaller than 1 K. The wafers were diced into 1 x 1 and 0.5 x 0.5 cm² chips for processing into the SNS edge junctions. Prior to photolithographic processing, the chips were ultrasonically cleaned for 30 minutes in trichloroethylene, acetone and ethanol to remove any residues left by the dicing process. In addition, the chips were plasma cleaned in a reactive ion etch (RIE) system for 30 minutes at 120 W with 150 mTorr O₂.

The YBCO/STO bilayer was prebaked at 130°C for one minute to remove adsorbed water vapor and coated with polyimide (CG285) spun at 5000 rpm for 40 seconds. The thickness of the polyimide layer was 500 nm. The polyimide-coated chip was partially cured at 130°C for five minutes on a hot plate. After the chip was allowed to cool, a 1.5 μm thick layer of 1518 photoresist was spun onto the chip at 5000 rpm for 40 seconds and prebaked at 95°C for 2 minutes. The

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resisted edge bead was removed with a 3 minute exposure, development in AZ developer diluted 1:1 in water, rinse and drying, in dry N_2 gas. The exposure of the chips was performed with a 350 nm UV flux of 10 mW/cm^2 through chrome contact masks. The base-electrode pattern was then exposed for one minute and developed in the AZ (1:1) solution. The 1518 resist pattern was then baked at 130°C for five minutes, hot enough to cause the edges of the resist to flow. Atomic Force Microscope (AFM) studies of the reflowed 1518 edges indicated that the edge angle, measured from the substrate plane, is -20° for line widths exceeding 5 μm .

The patterned chips were then mounted on a 1 mm-thick stainless-steel plate using Ag paint for milling using a 500 eV, 1 mA/cm^2 Ar⁺ ion beam at an angle 45° from normal incidence. (Fig. 1). The plate was rotated at 35 rpm using a DC motor mounted in the vacuum system. Typically, 17 minutes was required to etch through the YBCO, SrTiO_3 , and polyimide films; the total etching time was 20 minutes. Note that no cooling was provided to the sample during the entire ion-mill etch.

Following milling, the substrate was removed from the plate and excess Ag paint scrapped off of back. The sample was then placed in the RIE system for a 150 mTorr-O_2 plasma clean at 120 W for 30 minutes. This removed most of the cross-linked photoresist and polyimide. The sample was then ultrasonically cleaned in methylene chloride, acetone and ethanol for 10 minutes each. Methylene chloride is a solvent for polyimide CG285 and effectively lifts off any remaining moss-linked 1518 photoresist.

An AFM image of a YBCO/STO edge prepared with the polyimide-passivated reflowed resist is shown in Fig. 2. The edge is seen to be smooth and clear of large scale defects. The angles of the YBCO and SrTiO_3 portions of the edge are 17° and 10° , respectively, from the plane of the substrate. These separate angles presumably result from differences in milling rate.

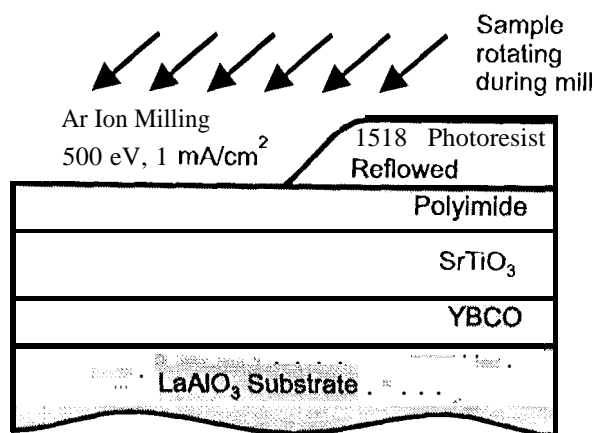


Fig. 1. - Cross section of the SNS edge junction structure during tapered edge formation. The polyimide passivation layer used below the reflowed-resist milling mask is unique to this work. The sample was not actively cooled during ion milling.

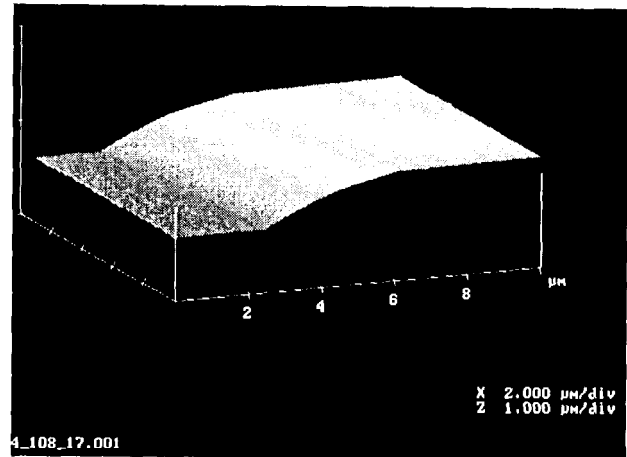


Fig. 2. - AFM image of a YBCO/STO edge formed by milling, the bilayer film coated with a polyimide passivation layer using a **rcfl()** vcd-resist mask. The sample holder was rotated but not actively cooled during ion milling.

Following the edge formation step, the YBCCO normal interlayer, $\text{YBa}_{1.95}\text{La}_{0.05}\text{Cu}_3\text{O}_{7-\delta}$ (YBICO) superconducting counterelectrode, and Au contact layer were deposited. The sample was mounted onto a Haines alloy plate with Ag paint and introduced into the load lock of a pulsed laser deposition (PLD) system containing a 5 cm-diameter ion source. The chip surface was spatter-cleaned in the load-lock chamber with a 100 eV, 1 mA/cm^2 ion beam of Ar and O_2 . The Ar partial pressure was 1.5×10^{-4} ; total pressure was 2.0×10^{-4} Torr. Etch time was five minutes, removing an estimated 5 nm of YBCO or SrTiO_3 .

The samples were then transferred *in situ* to the main deposition chamber. Prior to the deposition of the YBCCO N-layer and the YBICO counterelectrode films, the substrate was heated to 820°C in 200 mTorr O_2 for 20 minutes. The YBCCO and YBICO films were then deposited by PLD (fluence of 1.6 J/cm^2 at $\lambda = 248 \text{ nm}$). Substrate temperature was controlled by a thermocouple and measured with an optical pyrometer using heavily-doped Si chips mounted close to the device chips. During film deposition the laser spot was scanned over the 5 cm-diameter targets in a vertical direction and the substrate was oscillated $\pm 1 \text{ cm}$ in horizontal direction. This produced films with total thickness variation less than 10 % over a $1 \times 1 \text{ cm}^2$ area. Following film deposition, the chip was cooled at 3°C/min , to room temperature in 500 Torr of O_2 . Following this anneal step, a 100 nm-thick Au film was deposited by dc sputtering in 10 mTorr of Ar.

Edge SNS junction chips with YBCCO N-interlayer thicknesses of 0, 10, 46 and 93 nm were fabricated. The YBICO thickness was fixed at 200 nm. The YBCCO films had resistivities of $250 \mu\Omega\text{-cm}$ at 60 K and transition temperatures of 45 K. The transition temperature of the YBICO films were typically 88 K.

The chips were further processed to define base-electrode contacts and to define the counter-electrode bridges. SNS

device widths were 1.25 - 10 μm . We have previously reported this part of the edge SNS device fabrication[9].

B. Edge Junctions without Polyimide Passivation

Control devices were also prepared without the polyimide passivation layer under the resist. (The same 1518 photoresist and base-electrode patterning were used.) However, these chips were ion milled in an 10 cm-diameter system with a water-cooled, tiltable, rotating sample stage. Note that edges prepared with 1518 masks only and no water cooling were not acceptable for use in edge SNS devices. The chips were mounted on the sample stage with Ag grease. This system has a lower energy limit of 1000 eV, therefore the current density was lowered to $<0.2 \text{ mA/cm}^2$ to reduce damage due to the higher ion energy. A milling time of 45 minutes was required to clear the chips to the substrate. Devices with YBCO interlayer thicknesses of 0, 10, 20 and 30 nm were fabricated. The devices were completed with the process described above.

An AFM image of a YBCO/STO edge produced with this 1518 reflow process is shown in Fig. 3. The edge is smooth and clear of large scale defects, similar the edge shown in Fig. 2 produced by the polyimide passivation process. The YBCO edge angle of the edge is 13° from the plane of the substrate and the STO edge angle is 7° . Notably different between the two AFM images (Figs. 2 and 3) is the smoothness outside of the area of the edge. The sample processed with the polyimide passivation is smoother than the sample processed with only the reflowed resist. We further note that subsequent Y1(X)-based layers grown on the chips processed with the polyimide passivation appear cleaner, by optical microscopy, than those without the passivation layer.

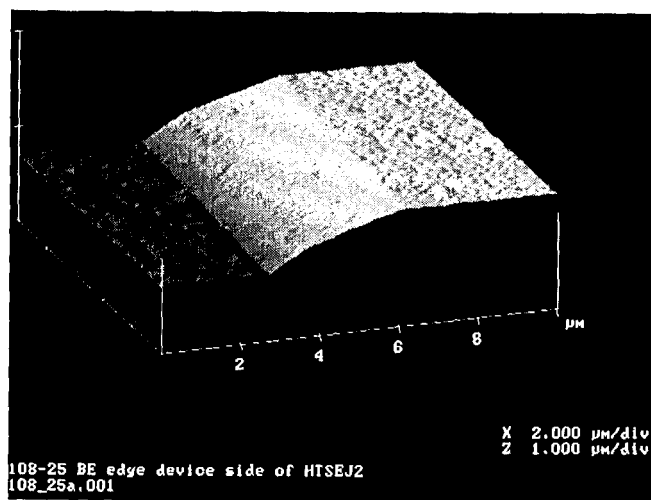


Fig. 3. - AFM image of a YBCO/STO film edge formed by ion-milling with a rf (lc) w.d-resist mask. The sample stage was water cooled during ion milling. Note the rougher surfaces than in Fig. 2.

11. ELECTRICAL MEASUREMENTS

Device chips were mounted in 28-pin ceramic chip packages and inserted into a magnetically shielded cryogenic probe equipped with a thermometer and resistive heater for DC measurements. The temperature was maintained by a controller. Four-wire measurements were performed with two Al wires ultrasonically bonded to the counter electrode of each device and two wires bonded to common base-electrode contacts. Current-voltage (IV) characteristics were obtained by current-biasing the devices using an audio function generator. The current, derived from a known series resistor, and the voltage signals from the device were fed into low-noise amplifiers, the outputs of which were fed into an analog-to-digital converter.

The IV characteristics of a typical SNS edge JJ at 77 K, with and without microwave radiation, are shown in Fig. 4. This device had a YBCCO interlayer thickness of 10 nm and was processed with the polyimide passivation layer. Similar IV characteristics were observed for junctions without the passivation layer. At 77 K, the critical-current density (J_c) of this device was $1.03 \times 10^4 \text{ A/cm}^2$, the resistance was 0.26Ω , and the critical current-resistance ($I_c R_n$) product was $48 \mu\text{V}$. The shape of the IV characteristic is qualitatively consistent with the resistively-shunted junction (RSJ) model with a small excess current. Well-defined Shapiro steps are evident under microwave irradiation.

For all devices fabricated, J_c was observed to decrease exponentially with increasing YBCCO thickness. For a given YBCCO thickness J_c was generally larger in devices fabricated with the polyimide passivation than in devices without it. Note, however, that the resistances of devices fabricated with the polyimide process were lower by about a factor of three than those of the devices fabricated with the reflowed resist mask alone, although all resistances were significantly higher than expected from YBCCO resistivity. This suggests a boundary or interface resistance in series with the normal interlayer. For YBCCO thicknesses of 10 nm, $I_c R_n$ values of

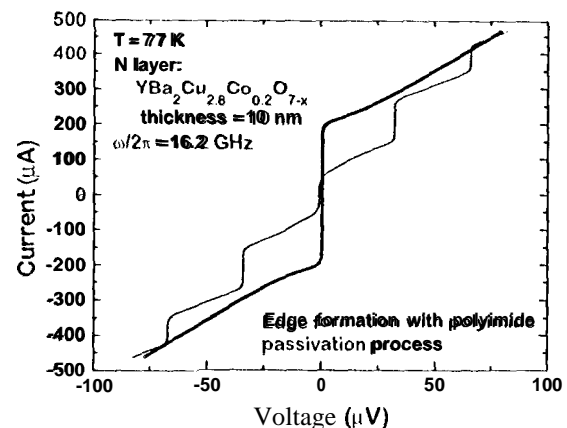


Fig. 4. - IV characteristics of an SNS device fabricated with the polyimide passivation process. The data were taken at 77 K.

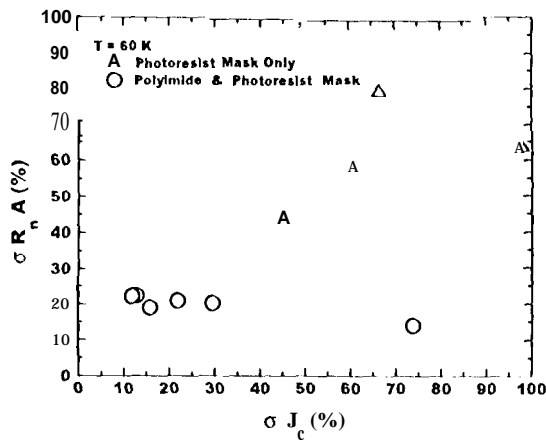


Fig. 5 - Device parameter spreads for each fabricated chip, with and without the polyimide passivation layer. The improved on-chip control with the polyimide process is clearly visible. The data were taken at 60 K and represent one standard deviation divided by the mean value expressed in percent.

150 - 800 μ V were observed at 60 K independent of the method used to fabricate the edge.

The most significant difference between the two processes is shown in Fig. 5, in which the on-chip spread in $R_n A$ (1σ) is plotted against the spread in J_c . The data were taken at 60 K. All data from ten devices measured on each fabricated chip are included. Clearly, device parameter spreads are more tightly controlled in the polyimide process than in the simple reflow process. Note that the 74% spread in J_c for the 93 nm-thick YBCCO polyimide-passivated chip is accounted for by the fact that the devices were just beginning to show an observable critical current at the measurement temperature. At 50 K, 1σ for J_c was 24%.

IV. CONCLUSION

We have fabricated SNS edge JJ's using a polyimide passivation layer underneath a reflowed-resist mask without actively cooling the chips during the ion-mill etch. This process produces devices with excellent IV characteristics and $I_c R_n$ values as high as 800 μ V at 60 K. Compared with chips fabricated without the passivation layer on-chip device parameter spreads were significantly reduced by the new process.

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REFERENCES

- [1] K.A. Delin and A. W. Kleinsasser, "Stationary properties of high critical temperature proximity effect Josephson junctions", *Supercond. Sci. Technol.* **9**, 227-269 (1996)
- [2] J. Gao, W.A.M. Aarnink, G.J. Gerritsma, and H. Rogalla, "Controlled preparation of all high- T_c SNS-type edge junctions and dc SQUIDs", *Physica C*, **171**, 126-130 (1990)
- [3] B.D. Hunt, M.C. Foote, and J. Bajuk, "All high- T_c edge-geometry weak links utilizing Y-Ba-Cu-O barrier layers", *Appl. Phys. Lett.* **59**, 982-984 (1991)
- [4] K. (bar), L. Antognazza, and M. Geballe, "Properties of $YBa_2Cu_3O_{7-x}/YBa_2Cu_3O_{7-x}$ edge junctions", *Appl. Phys. Lett.* **65**, 904-906 (1994)
- [5] B.D. Hunt, M. G. Forrester, J. Talvacchio, J.D. McCambridge, and R. M. Young, "High- T_c superconductor/normal metal/superconductor edge junctions and SQUIDs with integrated groundplanes", *Appl. Phys. Lett.* **68**, 3805-3807 (1996)
- [6] W.H. Mallison, S.J. Berkowitz, A.S. Hirahara, M.J. Neal, and K. (bar), "A multilayer $YBa_2Cu_3O_x$ Josephson junction process for digital circuit applications", *Appl. Phys. Lett.* **68**, 3808-3810 (1996)
- [7] D.M. Byrne, A.J. Brouns, F.C. Case, R.C. Tiberio, B.I. Whitehead, and E.D. Wolf, "Infrared mesh filters fabricated by electron-beam lithography", *J. Vac. Sci. Technol. B*, **3**, 26X (1985)
- [8] H.G. LeDuc, private communication
- [9] B.D. Hunt, M.C. Foote, W.T. Pike, J.B. Barner and R.P. Vasquez, "High- T_c edge-geometry SNS weak links on silicon-on-sapphire substrates", *Physica C* **230**, 141-152 (1994) and the references therein.

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